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Fukushima and Chernobyl nuclear accidents' environmental assessments and U.S. Hanford Site's waste management

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Abstract

This paper describes environmental clean-up activities at Fukushima Prefecture, Japan and a proposed strategic remediation plan, the Chernobyl remediation in Ukraine, and Hanford Site of the U.S. Department of Energy. For the Fukushima case, cesium is strongly adsorbed by fine soil, and it has mostly remained on the land surface. Thus, removal of a thin top surface of the contaminated soil is an effective environmental clean-up method. Chernobyl remediation generally aims to eliminate relevant radionuclide pathways to humans and the Hanford Site have been undergone both reducing contamination levels and cutting-off pathways to humans.

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1. Introduction

The magnitude-9 East Japan Great Earthquake and subsequent tsunami damaged northeastern Japan on March 11, 2011. The tsunami, in turn caused Fukushima Daiichi Nuclear Plant's reactor core melt accident that amounted to approximately 1/7 of the radionuclides released to the environment by the 1986 Chernobyl nuclear accident. These released radionuclides are mostly ^{131}I, ^{134}Cs and ^{137}Cs, with very small amounts of ^{89}Sr, ^{90}Sr, ^{238}Pu, and $^{239\text{-}240}$Pu [1]. Main long-life radionuclides affecting human health by the 1986 Chernobyl nuclear accident are ^{137}Cs, ^{90}Sr, and plutonium [2]. Although it is not due to an accident,

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defense nuclear activities of the Hanford Nuclear Reservation Site of the U.S. Department of Energy over a half century have contaminated the soil, groundwater and the Columbia River in and around the site [3]. The site located in the eastern Washington State has been undergoing extensive remediation activities over a decade.

Radionuclides released to the environment undergo the transport and fate processes through the following mechanisms:

(1) Transport of radionuclides by water and air movements.
(2) Transport, deposition and re-suspension of particulate radionuclides by soil and sediment movements.
(3) Adsorption and desorption causing radionuclide phase changes between dissolved and particulate forms.
(4) Radionuclide decay and production of daughter products.
(5) Radionuclide influx from and efflux to other environmental media (e.g., radionuclide transfer from air to surface soil, from surface soil to groundwater and surface water).

Because environmental remediation takes a great deal of time and resources to develop needed technologies and to implement them to clean the environment, a scientifically defensive decision making process must be implemented for the environmental remediation. This paper presents a proposed comprehensive remediation decision making methodology, and actual remediation activities performed at Fukushima, Chernobyl and Hanford cases.

2. Proposed comprehensive remediation decision making methodology

A proposed environmental remediation decision making methodology is to:

(1) Provide a scientifically valid comprehensive decision making process of environmental clean-up.
(2) Implement effective specific remediation activities in a given contaminated location.
(3) Decide the priority of the remediation activities.
(4) Promote public participation to remediation decision making.

This remediation decision making methodology is to be set up in a webpage and consists of the following five distribution maps covering the entire contaminated area:

(1) Distribution Map 1: Environmental parameter map.
(2) Distribution Map 2: Radionuclide transport parameter map.
(3) Distribution Map 3: Radionuclide migration map.
(4) Distribution Map 4: Remediation map.
(5) Distribution Map 5: Remediation priority map.

Distribution Map 1 is an environmental parameter map and consists of the following distribution maps which are layered on top of one another

(1) ^{134}Cs and ^{137} Cs concentration distribution.
(2) Population distribution.
(3) Geometry (mountain, plain, river, irrigation water way, etc.).
(4) Land use (residential area, farm field, dairy farmland, school, hospital, road, park, etc.).
(5) Soil and geological characteristics (soil chemistry, soil size distribution, etc.).
(6) Groundwater parameters (groundwater table height, hydraulic gradient, etc.).

Distribution Map 2 is a radionuclide transport parameter map and consists of

(1) Weather maps (rain, snow, air temperature, etc.).
(2) Radionuclide migration parameters
    (a) Land surface
    - land slope
    - soil erosion rate
    (b) Subsurface
    - porosity
    - hydraulic conductivity, etc

Distribution Map 3 is a radionuclide migration map and consists of

(1) Radionuclide migration rate.
    (a) Land surface
    - radionuclide adsorption (distribution coefficient)
    - overland runoff amount (rain and snowmelt)
    - soil erosion rate
    - radionuclide transport rate over the land surface
    (b) Subsurface
    - radionuclide adsorption (distribution coefficient)
    - radionuclide transport rate in unsaturated subsurface water
    - radionuclide transport rate in groundwater

Distribution Map 4 is to select appropriate remediation technology for each area shown in Distribution Map 1. It uses cesium remediation technology lists for contaminated soil, contaminated plants, agricultural water, groundwater, and surface water (river water and ocean water). A soil remediation technology list is shown in Table 1. For the contaminated water, zeolite and Prussian Blue are usually effective to remove dissolved cesium in water [4].

For each area shown in Distribution Map 1, Distribution Map 4 consists of

(1) Remediation area size and amount.
(2) Expected debris amounts needed to be treated and disposed .
(3) Performance and maturity of remediation technologies.
(4) Performance and maturity of waste treatment methods.
(5) Performance and maturity of waste disposal methods.

Distribution Map 5 is a remediation priority decision map and considers

(1) Environmental and human health risk assessment under.
    (a) Current condition
    (b) Future conditions due to radionuclide migration
(2) Achievability of securing disposal sites.
(3) Current and future land uses.
(4) Social and cost considerations.
(5) Others.

Table 1. General remediation technologies

Technology	Description	Comment
Excavation	Scrape upper soil layer and either wash soil or dispose	Effective, but remove valuable topsoil unless replaced with new or washed soil
Isolation	Engineered cover	Isolates contaminated materials and reduces exposure
Grouting	Inject grout material to entrap the radionuclides in a monolith	Isolates radionuclides, but restrict future land uses
In-situ leaching		Applicable to shallow soil. Excess leachate must be collected. Risk of uncontrolled mobilization. Effectiveness depends on soil characteristics
Physical and radiological soil separation	Separate soils with high concentrations from soils with low concentrations	Mature technologies. Effectiveness depends on soil characteristics
Ex-situ soil washing	Extract cesium from solids by washing with water or suitable extraction solutions	Effectives depends on soil characteristics

3. Fukushima nuclear accident

The East Japan Great Earthquake on March 11, 2011 occurred in the Pacific Ocean 69 km off northern Japan. It generated as high as 40 m high tsunami that hit the Japanese coast. A part of this tsunami came to the Fukushima Daiichi Nuclear Plant Site in Fukushima Prefecture in Japan, and caused the plant site-wide electric blackout. Without electricity, the plant lost its ability to supply water to cool nuclear fuels in Reactor Units 1, 2 and 3, resulting in nuclear core meltdowns. These core meltdowns induced hydrogen explosions in Units 1 and 3. Unit 4 also had a hydrogen explosion, despite the fact that Unit 4 was under scheduled maintenance at the time of the tsunami and all nuclear fuels were already moved from a reactor pressure vessel to spent fuel storage pool within Unit 4 reactor building. These three hydrogen explosions, together with venting air from Units 1, 2 and 3 released radionuclides, mostly ^{131}I, ^{134}Cs and ^{137}Cs into the air. These radionuclides with 3×10^{17} Becquerel (Bq) of radioactivity spewed into the atmosphere, were subsequently deposited on the land and ocean. Some radionuclides were also directly discharged into the Pacific Ocean from Fukushima Nuclear Plant. These releases contaminated the land, water, plant, and fish. To protect people from the this nuclear accident impacts, the Japanese Government evacuated over 70 000 people, who were living near the Fukushima Nuclear Power Plant.

A radionuclide half life of ^{131}I is 8 days. So it is already decayed out in the environment. Half lives of ^{134}Cs and ^{137}Cs are two and 30 years, respectively. Thus, they, especially ^{137}Cs will remain in the environment for sometime to come. There are many pathways for these radionuclides to affect people's health. They include direct radiation from the contaminated ground, drinking of contaminated water, and consumption of contaminated foods and soil. Table 2 shows the relatively conservative conversion factors from cesium concentration in Bq/m^3 to dosage in Sv through airborne pathways [3]. Because ^{137}Cs has a daughter product of ^{137}Ba, its effect is the sum of ^{137}Cs and ^{137}Ba effects, as shown in the bottom row.

Table 2. Conversion factors from cesium concentration in Bq/m3 to dosage in Sv through airborne pathways

Nuclide	Half-life	Inhalation/10^{-5}	Plume/10^{-6}	Ground/10^{-2}	Vegetable/10^{-2}	Milk/10^{-2}	Meat/10^{-2}	Soil/10^{-3}	Total/10^{-2}
^{134}Cs	2.062 y	9.9	2.0	4.2	1.3	3.4	4.3	0.6810	13
^{137}Cs	30.0 y	6.8	–	–	2.3	4.1	5.1	3.110	12
^{137}Ba	2.552 m	–	0.65	9.9	–	–	–	–	9.9
^{137}Cs		6.8	0.65	9.9	2.3	4.1	5.1	3.110	22.1

This table reveals that beside the direct radiation from the contaminated ground, consumption of contaminated foods, especially milk and meat are dominant radiation pathways to humans.

To protect people from the harmful radiation impacts, one can decontaminate the radioactively contaminated environment and/or eliminate the relevant pathways. This requires the determination of the radionuclide migration in the environment. The radionuclide transport and fate mechanisms are

(1) Transport of dissolved cesium by flowing water.

(2) Migration (transport, deposition and resuspension) of sediment—sorbed cesium by migration soil and sediment.

(3) Radionuclide adsorption to and desorption from soil and sediment.

(4) Radionuclide decay.

A portion of cesium is adsorbed by soil, but the amount of adsorption depends on soil characteristics, water quality, etc. [4]. Figures 1 and 2 show variations of cesium adsorption as a function of soil and water characteristics [5, 6]. They express cesium adsorption by a distribution function (see Eq. (1)) varying with soil and water characteristics.

$$K_d = \frac{C_p}{C_d} \tag{1}$$

where C_d is the dissolved cesium concentration, C_p is the sediment—sorbed cesium concentration, K_d is the distribution coefficient.

As shown in Fig. 1, the cesium distribution coefficient decreases with sodium concentration. This is also true with potassium. This is because cesium competes with sodium and potassium to attached to soil. This behavior may be used to reduce cesium concentration attached to soil by injecting sodium and potassium solutions over the contaminated soil. Moreover this figure also indicates the large cesium adsorption variation with clay minerals, having the strongest affinity to clay mineral of illite, while cesium is adsorbed the least to kaolinite.

Figure 2 indicates that cesium is less adsorbed by sediment, as chlorine increases. This implies that a portion of cesium adsorbed on sediment in a river (a freshwater) desorbs when the cesium—sorbed sediment reaches sea water, thus potentially increasing a dissolved cesium concentration in the saline water environment.

To protect people from harmful radiation effects of the Fukushima nuclear accident, the Japanese Government has been remediating the contaminated environment in mainly the following ways

(1) Remove
   (a) top several cm of contaminated soil.
   (b) weeds and other groundcover.
   (c) fallen leaves around residential areas.

(2) Cut low－hanging tree branches and remove moss on a tree trunk.

(3) Wash roof, wall and concrete road with high－pressure water jet.

(4) Sand－blast concrete road surface.
(5) Remove dissolved cesium in a swimming pool by zeolite.
(6) Impose institutional control
  (a) Evacuate people within a 20 km zone and other areas with 20 mSv/year or greater radiation exposure.
  (b) Prohibit consumption of foods with 100 Bq/kg or higher radionuclide concentration.

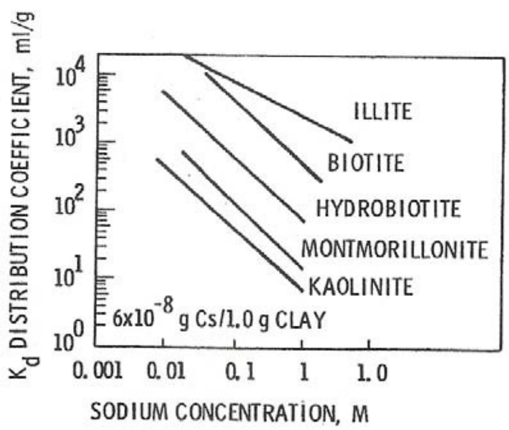

Fig. 1. Cesium adsorption with sodium and Clay minerals

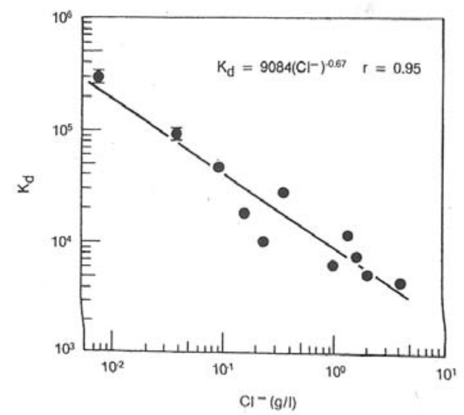

Fig. 2. Variation of cesium adsorption with chlorine

Remaining issues of Fukushima environmental remediation are
(1) Need to collect large volumes of contaminated soils, leaves and plants
(2) Waste storage
(3) Waste volume reduction
(4) Waste treatment of cesium contaminants
  (a) Removal of cesium from soil
    - difficult in a large scale operation
  (b) Removal of cesium from water
    - available, e.g., use of zeolite and Prussian Blue
  (c) Treatment of secondary waste
(5) Waste disposal
(6) Evaluation of future cesium migration and accumulation in solid and water

4. Chernobyl nuclear accident

During 10 days starting from April 26, 1986, the Chernobyl Nuclear Plant accident spewed 1.85×10^{18} Bq of radionuclides into air. This amount is approximately six times that of the Hiroshima atomic bomb [7]. The nuclear plant is located approximately 120 km from Kiev, Ukraine along the Pripyat River, a tributary of the Dnieper River that discharges its water into the Black Sea. These radionuclides released from Unit 4 of the Chernobyl Nuclear Plant subsequently deposited on the land surface, contaminating the environment. Many of these deposited radionuclides reached to the Dnieper River and its tributary, Pripyat River. The Dnieper River provides 80% of the freshwater needs of Ukraine. This water is drinking water of Kiev, the capital of Ukraine, and is extensively used as irrigation water, through which 20 million Ukrainians have been contaminated through consumption of foods that are produced with the irrigation water. Main radionuclides that have been inversely affecting people and the environment are shorter lived ^{131}I and longer lived ^{137}Cs, ^{90}Sr and $^{239, 240, 241}$Pu.

The former Soviet Union and Ukraine have conducted various countermeasures to protect people. These include

(1) Decontamination of contaminated water with the use of zeolite, bentonite, activated charcoal, and mineral fertilizers (e.g., K, P, N, lime).
(2) Physical methods to retard radionuclide migration.
(3) Providing cleaner water supplies, e.g., new wells and more use of the Desna River.
(4) Administration control, e.g., 30 km evacuation zone, ban of fishing and forest use.
(5) Better planning and the environment and health assessment.

The main remediation efforts have been to eliminate or reduce radionuclide migration from contaminated areas to the cleaner areas. Two specific examples are construction of a dike along the Pripyat River, and the second example is the construction of the new safe confinement (NSC) over the current Chernobyl shelter, formally called the "Sarcophagus".

A Pripyat River floodplain across the Chernobyl Nuclear Plant is one of the most contaminated areas by the Chernobyl accident and contains 3×10^{14} Bq of ^{90}Sr. A flooding over this area transports some of ^{90}Sr into the Pripyat River, thus also to the Dnieper River, supplying almost a half of ^{90}Sr moving through the Dnieper River. To build a dike along the Pripyat River to block off the flooding water to get into this floodplain is a viable remediation. The 4 year flood results in the highest ^{90}Sr concentrations in the river by just barely covering this 13 km by 3.8 km floodplain. Figure 3 and 4 show predictions of the FETRA code [2] under the 4 year flood without and with the new dike. As shown in these figures, construction of the new dike reduced ^{90}Sr Concentrations in the Pripyat River by almost half, as field data indicated.

Although the current Chernobyl Shelter has been standing there for over 25 years, the current Chernobyl Shelter is an temporary structure. Thus it has a danger of collapsing that would disperse a large amount of radionuclides contained in the Chernobyl Shelter into the air. To reduce the impact of its possible collapse, 270 m wide, 144 m long, the 100 m high, movable NSC is being constructed over the Chernobyl Shelter. The construction of the NSC would reduce the impacts of the Chernobyl Shelter collapse by approximately 95% by decreasing the radionuclide migration through atmosphere, groundwater and the Pripyat River [2].

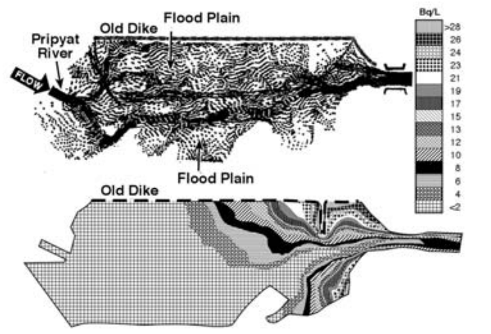

Fig. 3. Predicted flow and 90Sr concentrations without the New Dike

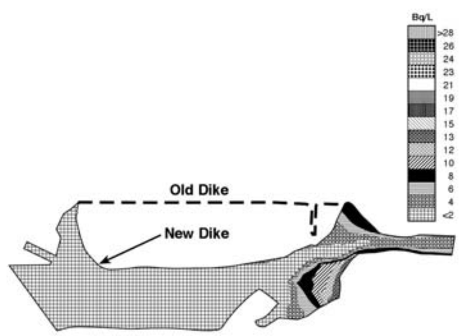

Fig. 4. Predicted 90Sr concentrations with a new dike

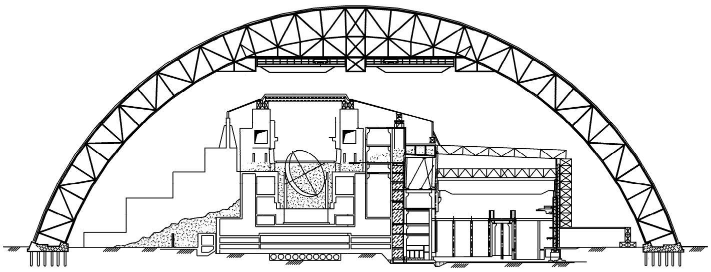

Fig. 5. Chernobyl new safe confinement

5. Hanford site

U.S. Department of Energy's Hanford Site is located along the Columbia River in southeastern Washington State. It was a part of the Manhattan Project. Its main mission had been to produce nuclear fuels, spent nuclear fuels in nuclear reactors, and reprocess the spent fuels to produce plutonium for nuclear weapons. The Hanford Site had nine nuclear reactors along the Columbia River, including the world first full scale nuclear reactor, B Plant. The Columbia River water was used as a once though cooling water, and the water coming out of the reactors were released to the environment. The spent nuclear fuels out of the reactors were reprocessed to obtain plutonium, and generated 200 000 m^3 of waste containing 7×10^{18} Bq of radioactivity. The wastes are a mixture of high-level (HLW) and low-level (LLW) radioactive and transuranic (TRU) waste and chemicals. These waste have been stored in the 177 of mostly 400 000 m^3 underground tanks. Many of these tanks are beyond their design lives. Sixty seven of these underground tanks were leaking or suspected of leaking, thus threatening to contaminate the nearby Columbia River through the groundwater seepage. These Hanford activities resulted in contamination of the Hanford soil and groundwater, and the Columbia River. For example, 170 km^2 of contaminated groundwater exists under the Hanford Site. These environmental contaminations pollute plants, terrestrial and aquatic biota in and outside of the Hanford Site, thus can potentially affects the human health.

To protect the environment and human health, the Hanford Site undergoes extensive remediation activities, including [8]

(1) Decommissioning and long-term storing of the rectors.
(2) Dismantling of various nuclear facilities.
(3) Removal of waste from the underground tanks.
(4) Cleaning up the Hanford contaminated soil and groundwater.
(5) Treatment of radioactive and chemical wastes.
(6) Storage of waste.
(7) Disposal of waste and contaminated water and soil.

The waste management work at the site includes to

(1) Transfer waste stored in 149 leaky-prone single-shell (single-layer) underground tanks (SSTs) to 28 safer double-shell (double-layer) tanks (DSTs).
(2) Constructing the Hanford Tank waste treatment and immobilization plant (WTP) to solidify the tank waste.
(3) Transfer tank waste stored in DSTs to WTP.

(4) Chemically prepare waste for waste solidification for both HLW and low activity waste (LAW; a low radiation HLW) at WTP.

(5) Vitrify (glassify) HLW and LAW.

(6) Dispose LAW in the on-site LLW disposal facility called the environmental restoration disposal facility (ERDF) and store vitrified HLW until final disposal off site.

The tank waste is multiphase, multi-component, high-ionic strength, and highly basic mixtures of liquids, solids, and, in some cases, gases. As shown in Fig. 6, Three-hundred-horsepower mixer pumps in the 4 000 m^3 DSTs to stir the radioactive sludge and supernatant liquid (and, in many cases, an added solvent) so the waste can be retrieved from the tanks for subsequent treatment and disposal at WTP. During the retrieval operation, complex interactions occur between waste mixing and chemical reactions. Main chemical reactions of the water dissolving tank waste occurs among chemical species, as shown in Fig. 7. This figure shows main chemical species of the water resolvable tank waste. It also shows prediction results of solid resolution after the tank waste is mixed with water by mixer pumps shown in Fig. 7, as predicted by the three-dimensional reactive transport code, ARIAL [9].

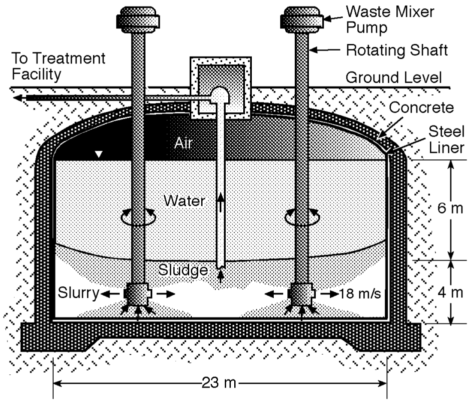

Fig. 6. Hanford tank waste retrieval

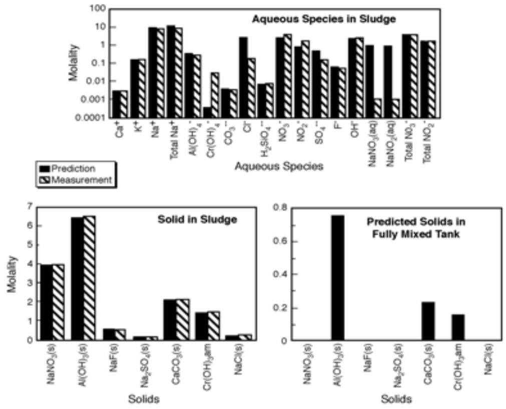

Fig. 7. Measured and predicted waste chemistry

As a part of the environmental clean up, the 54 ha ERDF was constructed in the Hanford Site as a LLW and hazardous and mixed waste disposal site. It has multiple protective layers to prevent waste leaking, and monitors and collect any leakage, if any. It will be expanded as needed. So far, eleven million tons of contaminated soil in the Hanford Site had been removed and were disposed in the ERDF.

For groundwater decontamination, various methods are being used. They include the "pump and treat". This method withdraws the contaminated groundwater from an aquifer under the Hanford Site, treat it to eliminate ^{99}Tc and some toxic chemicals, and injected the decontaminated water back to the aquifer at different locations. Although it takes time, it is effective to remediate the groundwater contaminated by radionuclides and chemicals having the small affinity to soil (small distribution coefficients). However, it is not effective to radionuclides, e.g., cesium and plutonium having strong affinity to soil. But the "pump and treat" can be effective even to these radionuclides to change the direction and speed of their migration to avoid reaching to critical areas by changing the hydraulic gradient of the groundwater.

The Hanford Site uses willow trees to remove ^{90}Sr from the groundwater. Another way to remove ^{90}Sr from the groundwater is to inject solutions of calcium and phosphorus to an aquifer to form apatite. When the groundwater containing ^{90}Sr encounters the apatite, calcium in apatite exchanges with ^{90}Sr in the groundwater to trap ^{90}Sr as a part of apatite. This process removes ^{90}Sr from the groundwater.

6. Summary and conclusions

This paper discussed environmental remediation activities as countermeasures of the Fukushima nuclear accident, Chernobyl nuclear accident, and the Hanford site remediation through decommissioning, decontamination and waste management. Once the environment is contaminated, it takes many specialized technologies, and large amounts of resources and duration to clean-up the environment. To protect humans from harmful effects of radionuclides released to the environment, one needs to holistically and systematically consider the environmental remediation of the contaminated sites and eliminating relevant pathways to humans.

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